

CLAIMS

1. A production method of a DDR type zeolite membrane, characterized in that a DDR type zeolite membrane is formed by carrying out hydrothermal synthesis with using a raw material solution having a containing ratio of 1-adamantanamine to silica (1-adamantanamine/SiO₂) of a molar ratio of 0.03 to 0.4, a containing ratio of water to the silica (water/SiO₂) in a molar ratio of 20 to 500, and a containing ratio of ethylenediamine to the 1-adamantanamine (ethylenediamine/1-adamantanamine) in a molar ratio of 5 to 32; and a DDR type zeolite powder to be a seed crystal.
2. The production method of a DDR type zeolite membrane according to claim 1, wherein said raw material solution has a containing ratio of said 1-adamantanamine to said silica (1-adamantanamine/SiO₂) of 0.05 to 0.25 in a molar ratio, a containing ratio of said water to said silica (water/SiO₂) of 28 to 220 in a molar ratio, and a containing ratio of said ethylenediamine to said 1-adamantanamine (ethylenediamine/1-adamantanamine) of 8 to 24 in a molar ratio.
3. The production method of a DDR type zeolite membrane according to claim 1 or 2, wherein said raw material solution is prepared by dissolving said 1-adamantanamine in said ethylenediamine to prepare a 1-adamantanamine solution, and then mixing said 1-adamantanamine solution with said silica sol solution containing silica.

4. The production method of a DDR type zeolite membrane according to any one of claims 1 to 3, wherein hydrothermal synthesis is performed at 130°C to 200°C.
5. The production method of a DDR type zeolite membrane according to any one of claims 1 to 4, wherein said DDR type zeolite powder is dispersed in said raw material solution.
6. The production method of a DDR type zeolite membrane according to any one of claims 1 to 5, wherein said DDR type zeolite membrane is formed on a porous substrate.
- 10 7. The production method of a DDR type zeolite membrane according to any one of claims 1 to 4, wherein said DDR type zeolite powder is deposited on a porous substrate, and said raw material solution is brought into contact with said porous substrate to form said DDR type zeolite membrane on said porous substrate.
- 15 8. The production method of a DDR type zeolite membrane according to claim 6 or 7, wherein a thickness of said DDR type zeolite membrane formed on said porous substrate is 0.1 to 50 µm.
- 20 9. The production method of a DDR type zeolite membrane according to any one of claims 6 to 8, wherein said porous substrate is in the form of a plate, a cylinder, a honeycomb, or a monolith having a plurality of cylindrical tubes integrated.
- 25 10. A DDR type zeolite membrane, characterized in that it is formed as a membrane on a substrate and its main component is silica, and that each single gas permeance at room

temperature and 100°C are different, respectively among at least two types of gases selected from a group consisting of carbon dioxide (CO₂), hydrogen (H₂), oxygen (O₂), nitrogen (N₂), methane (CH₄), normal butane (n-C₄H₁₀), isobutane (i-C₄H₁₀), sulfur hexafluoride (SF₆), ethane (C₂H₆), ethylene (C₂H₄), propane (C₃H₈), propylene (C₃H₆), carbon monoxide (CO), and nitrogen monoxide (NO).

11. The DDR type zeolite membrane according to claim 10, wherein a gas permeance of carbon dioxide (CO₂) at room 10 temperature is 1.0×10^{-9} (mol·m⁻²·s⁻¹·Pa⁻¹) or more.

12. The DDR type zeolite membrane according to claim 10, wherein a gas permeance of carbon dioxide (CO₂) at 100°C is 5.0×10^{-10} (mol·m⁻²·s⁻¹·Pa⁻¹) or more.

13. The DDR type zeolite membrane according to any one of 15 claims 10 to 12, wherein a separation factor of CO₂/CH₄ in a mixed gas containing carbon dioxide (CO₂) and methane (CH₄) in an equimolar amount is 2 or more at room temperature and 100°C.

14. The DDR type zeolite membrane according to claim 10, 20 wherein each value of a ratio of a single gas permeance of carbon dioxide (CO₂) at room temperature and 100°C to a single gas permeance of any one of hydrogen (H₂), oxygen (O₂), nitrogen (N₂), methane (CH₄), normal butane (n-C₄H₁₀), isobutane (i-C₄H₁₀), and sulfur hexafluoride (SF₆) at room 25 temperature and 100°C is 2 or more.

15. The DDR type zeolite membrane according to claim 14, wherein a value of a ratio of a single gas permeance of

hydrogen (H_2) at room temperature and 100°C to a single gas permeance of any one of oxygen (O_2), nitrogen (N_2), methane (CH_4), normal butane ($n-C_4H_{10}$), isobutane ($i-C_4H_{10}$), and sulfur hexafluoride (SF_6) at room temperature and 100°C is 2 or more.

5 16. The DDR type zeolite membrane according to claim 14 or 15, wherein each value of a ratio of a single gas permeance of oxygen (O_2) at room temperature and 100°C to a single gas permeance of any one of nitrogen (N_2), methane (CH_4), normal butane ($n-C_4H_{10}$), isobutane ($i-C_4H_{10}$), and sulfur hexafluoride
10 (SF_6) at room temperature and 100°C is 1.1 or more.

17. The DDR type zeolite membrane according to any one of claims 14 to 16, wherein each value of a ratio of a single gas permeance of nitrogen (N_2) at room temperature and 100°C to a single gas permeance of any one of methane (CH_4), normal
15 butane ($n-C_4H_{10}$), isobutane ($i-C_4H_{10}$), and sulfur hexafluoride (SF_6) at room temperature and 100°C is 2 or more.

18. The DDR type zeolite membrane according to any one of claims 14 to 17, wherein each value of a ratio of a single gas permeance of methane (CH_4) at room temperature and 100°C to a single gas permeance of any one of normal butane ($n-C_4H_{10}$), isobutane ($i-C_4H_{10}$), and sulfur hexafluoride (SF_6) at room
20 temperature and 100°C is 2 or more.

19. The DDR type zeolite membrane according to any one of claims 14 to 18, wherein each value of a ratio of a single gas permeance of normal butane ($n-C_4H_{10}$) at room temperature and 100°C to a single gas permeance of isobutane ($i-C_4H_{10}$) or

sulfur hexafluoride (SF_6) at room temperature and 100°C is 1.1 or more.

20. The DDR type zeolite membrane according to any one of claims 14 to 19, wherein each value of a ratio of a single gas permeance of isobutane ($i-C_4H_{10}$) at room temperature and 100°C to a single gas permeance of sulfur hexafluoride (SF_6) at room temperature and 100°C is 1.1 or more.

21. The DDR type zeolite membrane according to claim 10, wherein each separation factor of H_2/CH_4 in a mixed gas containing hydrogen (H_2) and methane (CH_4) in an equimolar amount at room temperature and 100°C is 2 or more.

22. The DDR type zeolite membrane according to claim 10, wherein each separation factor of C_2H_4/C_2H_6 in a mixed gas containing ethylene (C_2H_4) and ethane (C_2H_6) in an equimolar amount at room temperature and 100°C is 1.5 or more.

23. The DDR type zeolite membrane according to claim 10, wherein each separation factor of O_2/N_2 in the air at room temperature and 100°C is 1.5 or more.

24. A gas separation method for separating at least one type of gas component from a mixed gas containing at least two types of gas components selected from a group consisting of carbon dioxide (CO_2), hydrogen (H_2), oxygen (O_2), nitrogen (N_2), methane (CH_4), normal butane ($n-C_4H_{10}$), isobutane ($i-C_4H_{10}$), sulfur hexafluoride (SF_6), ethane (C_2H_6), ethylene (C_2H_4), propane (C_3H_8), propylene (C_3H_6), carbon monoxide (CO), and nitrogen monoxide (NO), using a DDR type zeolite membrane according to any one of claims 10 to 23.

25. The gas separation method according to claim 24, wherein carbon dioxide (CO₂) is selectively separated from a mixed gas containing carbon dioxide (CO₂) and methane (CH₄).

26. A gas separation apparatus comprising a DDR type zeolite membrane according to any one of claims 10 to 23 in order to separate at least one type of gas component from a mixed gas containing at least two types of gas components selected from a group consisting of carbon dioxide (CO₂), hydrogen (H₂), oxygen (O₂), nitrogen (N₂), methane (CH₄), normal butane (n-C₄H₁₀), isobutane (i-C₄H₁₀), sulfur hexafluoride (SF₆), ethane (C₂H₆), ethylene (C₂H₄), propane (C₃H₈), propylene (C₃H₆), carbon monoxide (CO), and nitrogen monoxide (NO).

27. The gas separation apparatus according to claim 26, wherein the gas separation apparatus selectively separates carbon dioxide (CO₂) from a mixed gas containing carbon dioxide and methane (CH₄).

28. A DDR type zeolite membrane composite, characterized by being provided with a porous substrate, and a DDR type zeolite layer deposited within pores of substrate and having a thickness 5 to 50 times of a mean pore diameter of the porous substrate; said DDR zeolite layer composed of a DDR type zeolite having been disposed within pores of at least one surface of the porous substrate.

29. The DDR type zeolite membrane composite according to claim 28, further comprising a DDR type zeolite layer deposited outside of the substrate, which is made of a DDR type zeolite and has a thickness of 30 µm or less, on a surface

of said porous substrate on which said DDR type zeolite layer deposited within pores of substrate is disposed.

30. The DDR type zeolite membrane composite according to claim 28 or 29, wherein a mean pore diameter of said porous 5 substrate is 0.05 to 10 μm .

31. A production method of a DDR type zeolite membrane composite, characterized by forming a raw material solution having a mixing ratio of 1-adamantanamine to silica (1-adamantanamine (mol)/silica (mol)) of 0.03 to 0.4, and a 10 mixing ratio of water to silica (water (mol)/silica (mol)) of 20 to 500, immersing a porous substrate in said obtained raw material solution for hydrothermal synthesis, thereby forming a DDR type zeolite layer deposited within pores of substrate having a thickness of 5 to 50 times of a mean pore 15 diameter of said substrate, and being formed from a DDR type zeolite, which is formed within pores of at least one surface of said porous substrate.

32. The production method of a DDR type zeolite membrane composite according to claim 31, wherein a DDR type zeolite 20 layer deposited outside of the substrate having a thickness of 30 μm or less, being formed from a DDR type zeolite on a surface of the porous substrate, on which the DDR type zeolite layer deposited within pores of substrate is disposed.

25 33. The production method of a DDR type zeolite membrane composite according to claim 31 or 32, wherein said porous substrate has a mean pore diameter of 0.05 to 10 μm .

34. The production method of a DDR type zeolite membrane composite according to any one of claims 31 to 33, wherein said hydrothermal synthesis is performed at 130°C to 200°C.

35. The production method of a DDR type zeolite membrane composite according to any one of claims 31 to 34, wherein 5 said raw material solution further contains a DDR type zeolite powder to be a seed crystal.

36. The production method of a DDR type zeolite membrane composite according to any one of claims 31 to 34, wherein 10 a DDR type zeolite powder to be a seed crystal is deposited on surface of said porous substrate to be immersed in said raw material solution.